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Smoothening Transition of Rough Surfactant Surfaces

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We propose a model for surfaces in mixtures of oil, water, and surfactants with strong electric dipoles. The dipole interactions give rise to a non-local interaction with a *negative stiffness* between surface elements. We show that, for large space dimension D, this model has a phase transition from *rough* to *smooth* surfaces. Contrary to models with a simple quadratic curvature energy of a positive sign, which always have a finite persistence length typical for rough surfaces, the smooth surfaces in our model exhibit *long-range* correlations of a generalized antiferromagnetic type. These correlations might be related to the recently observed "egg carton" superstructure of membranes.

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Models of random surfaces play an important role in chemical physics, biophysics, and particle physics. The statistical mechanics of such models [1] is usually discussed on the basis of a phenomenological Hamiltonian which contains a surface tension and a positive extrinsic curvature stiffness [2]. Freely suspended vesicles formed from such membranes have a vanishing tension, and their shapes are determined by a positive curvature energy [3]. Such a curvature energy is known to have only a finite persistence length ξ [4,5,6,7]. The correlation functions of the tangent vectors of the surface fall off like $e^{-|x|/\xi}$ signaling a rough surface [4,8] on scales larger than ξ. For biomembranes, this limitation is of no practical importance, since their persistence lengths are much larger than the size of the vesicles. This is not true, however, for layers of surfactants between oil and water, or for double layers in water alone. Their persistence lengths are of the order of a few hundred Å. Beyond this distance, membrane fluctuations are governed only by tension. This phenomenon is called *spontaneous generation of tension* [9]. Thus, in these models, the tension can never disappear completely. Experimentally, however, it is quite easy to generate large smooth interfaces. In fact, this is done industrially during the tertiary recovery of oil from microemulsions. The fractal surfactant interfaces between oil and water are smoothened by the addition of salt. This produces a desired phase separation in which the oil floats on top of a single interface. The purpose of this note is to point out a mechanism which may explain this.

Some years ago it was shown that the curvature stiffness of surfactants at the interfaces between oil and water [10] receives a significant contribution from the electric fields created by layers and bilayers of surface charge [11]. These surfactants possess electric dipoles which align along the normals to the interface. The dipoles can be quite large [12]. The range of the electric fields can be varied by the addition of salt. In a field theoretic description, the dipoles interact with an electric potential of a finite mass μ which is equal to the inverse Debye screening length, thus being directly governed by the salt concentration n [13]:

$$\mu = \sqrt{\frac{8\pi e^2 n}{\epsilon k_B T}} \ , \tag{1}$$

where k_B is the Boltzmann constant, e the electron charge, and ϵ the dielectric constant.

In our model we shall ignore the curvature stiffness generated by the mechanical properties of the surfactant molecules, or by the van der Waals forces [14]. Thus we consider only surfactants with large dipole moments, where the other forces can be ignored, and curvature stiffness is *completely* due to the dipole-dipole interactions.

The interaction between the dipole moments in three dimensions is similar to the tensor interaction between tangent tensors in the world surfaces of models of quark confinement, in which quarks are held together in the same way as magnetic charges would in a superconductor [15]. Similar models have been conjectured to describe the world surface between quarks in the confining phase of QCD [16]. A recent analysis in Refs. [17,18] indicates that such a model describes smooth surfaces, a fact which seems to be confirmed by recent Monte Carlo simulations [19].

By integrating out the electric potential one obtains a non-local interaction between normals to the surface. Unfortunetely, such an interaction is hard to treat analytically. For this reason we shall study the phase structure of a simplified version [20] of this model which retains the essential features of the original one, most notably a non-local interaction with a *negative stiffness*.

Our starting point is a Hamiltonian describing surfaces with tension, whose normal vectors are coupled to a fluctuating electric field $E_\mu(x) = \partial_\mu \phi(x)$:

$$H = r \int d^2\xi \sqrt{g} + \int d^3x \ \frac{1}{2} \ \left(\partial_\mu \phi \partial_\mu \phi + \mu^2 \phi^2 \right) + d \int d^2\xi \sqrt{g} \ \partial_\mu \phi \ N_\mu \ . \tag{2}$$

The surface is parametrized by the three functions $x_\mu(\xi_1, \xi_2)$ ($\mu = 1, 2, 3$). The parameter d denotes the (uniform) dipole density on the surface, and μ is the inverse Debye length (1). The intrinsic geometry of the surface resides in the induced metric

$$g_{ab} = \partial_a x_\mu \partial_b x_\mu \ , \qquad g = \det \ g_{ab} \ . \tag{3}$$

The extrinsic geometry is described by the normal vectors

$$N_\alpha = \frac{1}{2\sqrt{g}} \ \epsilon_{\alpha\mu\nu} \ \epsilon_{ab} \ \partial_a x_\mu \partial_b x_\nu \ , \qquad N_\alpha N_\alpha = 1 \ . \tag{4}$$

Here, greek letters refer to the embedding space, while latin letters denote surface coordinates. For simplicity we use units in which $\hbar = 1$, $c = 1$, and measure all momenta, energies, or distances in units of the microscopic cutoff Λ or its inverse, respectively. A natural cutoff is provided by the molecular size. We furthermore set $\Lambda = 1$.

The electric coupling of the surface can be simplified by writing it in terms of the trace $C = \mathrm{Tr} \ C_{ab}$ of the second fundamental form, which is equal to twice the mean curvature:

$$H = r \int d^2\xi \sqrt{g} + \int d^3x \ \frac{1}{2} \ \left(\partial_\mu \phi \partial_\mu \phi + \mu^2 \phi^2 \right) + d \int d^2\xi \sqrt{g} \ \phi C \ . \tag{5}$$

Integrating out the electric field, we obtain the non-local energy

$$H = r \int d^2\xi \sqrt{g} - \frac{d^2}{2} \int d^2\xi \int d^2\xi' \ \sqrt{g} C(\xi) \ Y\left(x(\xi) - x(\xi')\right) \ \sqrt{g'} C(\xi') \ , \tag{6}$$

where

$$Y(x) = \frac{\mathrm{e}^{-\mu|x|}}{4\pi|x|} \ . \tag{7}$$

is the Yukawa potential.

We then proceed by rewriting the non-local kernel in the embedding space as a corresponding non-local kernel on the surface. To this end we introduce a new local coordinate system around each point of the surface: ξ^1, ξ^2, χ^3. The coordinates ξ^1 and ξ^2 at $\chi^3 = 0$ describe the original surface. Together with the coordinate χ^3, they form a locally flat coordinate system orthogonal to the surface. The coordinate transformation is described by functions $x_\mu(\xi^1, \xi^2, \chi^3)$ which for $\chi^3 = 0$ coincide with the original parametrization $x_\mu(\xi^1, \xi^2)$ of the surface.

The Yukawa potential in (6) can be rewritten as

$$\int d\chi \int d\chi' \ \delta(\chi) \ \frac{1}{\mu^2 - \nabla^2} \ \delta^3\left(x(\xi, \chi) - x(\xi', \chi')\right) \ \delta(\chi') \ . \tag{8}$$

We can now use the transformation rules

$$\begin{aligned} \delta^3(x - x') &= \frac{1}{\sqrt{g}} \ \delta^2(\xi - \xi') \ \delta(\chi - \chi') \ , \\ \mu^2 - \nabla^2 &= M^2 - \nabla_\chi^2 \ , \\ M^2 &\equiv \mu^2 - \mathcal{D}^2 \ , \\ \mathcal{D}^2 &= \mathcal{D}^a \mathcal{D}_a = \frac{1}{\sqrt{g}} \ \partial_a g^{ab} \sqrt{g} \ \partial_b \ , \end{aligned} \tag{9}$$

where $\mathcal{D}_a$ denotes covariant derivatives along the surface, to rewrite the Yukawa potential as

$$\begin{aligned} Y\left(x(\xi) - x(\xi')\right) &= \int d\chi \ \ \delta(\chi) \frac{1}{M^2 - \nabla_\chi^2} \delta(\chi) \ \frac{1}{\sqrt{g}} \ \delta^2(\xi - \xi') \\ &= G\left(\mathcal{D}^2\right) \ \frac{1}{\sqrt{g}} \ \delta^2(\xi - \xi') \ , \end{aligned} \tag{10}$$

where G is formally defined by an expansion in powers of $(\mathcal{D}/\mu)^2$. In order to compute this, we first note that $(M^2 - \nabla_\chi^2)^{-1} \delta(\chi)$ is the Yukawa Green's function in one dimension,

and thus equal to $\exp(-M|r|)/2M$. The first delta function in (10) tells us then that we have to take this function at $r = 0$, implying that

$$G\left(\mathcal{D}^2\right) = \frac{1}{2\mu} \, \frac{1}{\sqrt{1 - \left(\mathcal{D}/\mu\right)^2}} \, . \tag{11}$$

Inserting this result into (6), we obtain

$$\begin{aligned} H &= r \int d^2\xi \sqrt{g} - \frac{d^2}{4\mu} \int d^2\xi \sqrt{g} \; C \frac{1}{\sqrt{1 - \left(\mathcal{D}/\mu\right)^2}} C \\ &= r \int d^2\xi \sqrt{g} - \frac{d^2}{4\mu} \int d^2\xi \sqrt{g} \; C^2 + \dots \; . \end{aligned} \tag{12}$$

This shows that dipole dominance leads to a non-local interaction with *negative* stiffness

$$\kappa = \frac{d^2}{4\mu} \, . \tag{13}$$

The formulation (12) is rather awkward for analytic computations due to the non-linear character of the trace C. For this reason we shall investigate a model [20], which is simpler to handle while embodying the most important features of (12), namely non-locality and negative stiffness. This model is formulated in terms of the tangent vectors $\mathcal{D}_a x_\mu$ and posseses the following Hamiltonian:

$$\beta H = \int d^2\xi \sqrt{g} \;\; g^{ab} \mathcal{D}_a x_\mu \; W\left(z, m, \mathcal{D}^2\right) \;\; \mathcal{D}_b x_\mu \; , \tag{14}$$

Here the index μ runs over $\mu = 1, \dots, D$, with D the dimension of the embedding space, which we keep variable from now on. The interaction kernel is

$$W(z, m, \mathcal{D}^2) = \frac{z}{1 - \mathcal{D}^2/m^2} \; , \quad \text{where} \;\; z = \frac{\beta r}{2} \; , \quad m = \frac{\sqrt{2r\mu}}{d} \; . \tag{15}$$

It has the expansion

$$W\left(z, m, \mathcal{D}^2\right) = z + s\mathcal{D}^2 + \dots \; , \tag{16}$$

with $s = z/m^2 = \beta\kappa$ being the reduced dimensionless stiffness parameter. These expansion terms correspond to the Hamiltonian

$$H = r \int d^2\xi \sqrt{g} + \kappa \int d^2\xi \sqrt{g} \;\; g^{ab} \mathcal{D}_a x_\mu \mathcal{D}^2 \mathcal{D}_b x_\mu + \dots \; , \tag{17}$$

which matches (up to boundary terms) (12) to this order.

This model can now be analyzed non-perturbatively by standard large-D techniques along the lines of Refs. [9,21,8] . To this end we introduce a dimensionless Lagrange multiplier matrix λ^{ab} to enforce the constraint $g_{ab} = \partial_a x_\mu \partial_b x_\mu$,

$$\beta H \to \beta H + \int d^2\xi \sqrt{g} \;\; \lambda^{ab} \left(\partial_a x_\mu \partial_b x_\mu - g_{ab} \right) \;\; . \tag{18}$$

We then parametrize the world-sheet in the Gauss map as

$$x_\mu(\xi) = \left(\xi_1, \xi_2, \phi^i(\xi) \right) \;\; , \qquad\qquad i = 3, \ldots, D \;\; , \tag{19}$$

where $-R_1/2 \leq \xi_1 \leq R_1/2$, $-R_2/2 \leq \xi^2 \leq R_2/2$ and $\phi^i(\xi)$ describe the $D-2$ transverse fluctuations. Then we search for an isotropic saddle point of the form

$$g_{ab} = \rho \;\delta_{ab} \;\; , \qquad\qquad \lambda^{ab} = \lambda \;g^{ab} \;\; , \tag{20}$$

for the metric and the Lagrange multiplier of infinite systems ($R_1, R_2 \to \infty$). At such a saddle point, we obtain a Hamiltonian

$$\beta H = 2 \int d^2\xi \;\; [z + \lambda(1-\rho)] + \int d^2\xi \;\partial_a \phi^i \left[\lambda + W\left(z, m, \mathcal{D}^2 \right) \right] \;\partial_a \phi^i \;\; . \tag{21}$$

Integrating out the transverse fluctuations, we obtain in the infinite-area limit the free energy

$$\beta F = 2 A_{\rm ext} \;\; (z + \lambda(1-\rho)) + A_{\rm ext} \frac{D-2}{8\pi^2} \rho \int d^2p \; \ln \left\{ p^2 \left[\lambda + W\left(z, m, p^2 \right) \right] \right\} \;\; , \tag{22}$$

where $A_{\rm ext} = R_1 R_2$ is the extrinsic, physical area. For large D, the fluctuations of λ and ρ are suppressed and these variables take their classical values, determined by the two saddle-point equations

$$\begin{aligned} \lambda &= \frac{D-2}{8\pi} \;\; \int_0^1 dp \; p \; \ln \left\{ p^2 \left[\lambda + W\left(z, m, p^2 \right) \right] \right\} \;\; , \\ \frac{\rho - 1}{\rho} &= \frac{D-2}{8\pi} \;\; \int_0^1 dp \; p \; \frac{1}{\lambda + W\left(z, m, p^2 \right)} \;\; , \end{aligned} \tag{23}$$

where we have introduced the ultraviolet cutoff by restricting the momentum integrations to $p < 1$. Inserting the first saddle-point equation into (22), we find

$$\beta F = 2 \left(z + \lambda \right) \;\; A_{\rm ext} \;\; , \tag{24}$$

showing that the tension is renormalized to

$$\alpha = 2(z + \lambda) \ . \tag{25}$$

The physics of our surfaces in the large-D limit is determined thus by the two saddle-point equations (23). The first of these equations requires the vanishing of the *saddle-function*

$$f(z, m, \lambda) \equiv \lambda - \frac{D-2}{8\pi} \int_0^1 dp \ p \ \ln \left\{ p^2 \left(\lambda + W \left[z, m, p^2 \right) \right] \right\} \ , \tag{26}$$

and determines the Lagrange multiplier λ as the solution of a transcendental equation. After this, the second saddle-point equation determines the metric via the equation

$$\rho = \frac{1}{f'(z, m, \lambda)} \ , \tag{27}$$

where a prime denotes the derivative with respect to λ.

The simple interaction (15) in (14) has the advantage that the saddle function (26) can be computed exactly:

$$\begin{aligned} f(z, m, \lambda) = \lambda &- \frac{D-2}{16\pi} \left\{ -1 - m^2 \ \ln \left(1 + \frac{1}{m^2} \right) - \frac{m^2(\lambda + z)}{\lambda} \ \ln \left(\frac{m^2(\lambda + z)}{1 + m^2} \right) \right\} \\ &- \frac{D-2}{16\pi} \left\{ \frac{m^2(\lambda + z) + \lambda}{\lambda} \ \ln \left(\lambda + \frac{z m^2}{1 + m^2} \right) \right\} \ . \end{aligned} \tag{28}$$

This function has the following limiting values:

$$\begin{aligned} \lim_{\lambda \to \infty} f(z, m, \lambda) &= \infty \ , \\ \lim_{\lambda \to \lambda_{\min}} &= -z m^2 - \frac{D-2}{16\pi} \left(-1 + \ln \ z m^2 \right) + \mathcal{O} \left(m^2 \ \ln \ m^2 \right) \ , \qquad m \ll 1 \ , \\ \lambda_{\min} &= \frac{-z m^2}{1 + m^2} \ . \end{aligned} \tag{29}$$

For z sufficiently large we have $\lim_{\lambda \to \lambda_{\min}}, < 0$ and there exists at least one solution to the saddle-point equation $f(z, e, \lambda) = 0$.

The derivative of the saddle-function f,

$$f'(z, e, \lambda) = 1 - \frac{D-2}{16\pi} \left\{ \frac{1}{\lambda} - \frac{z m^2}{\lambda^2} \left[\ln \left(1 + \frac{1}{m^2} \right) + \ln \frac{\lambda + \frac{z m^2}{1 + m^2}}{\lambda + \frac{z m^2}{m^2}} \right] \right\} \ , \tag{30}$$

determines the metric element ρ via (27). Given that

$$\begin{aligned} \lim_{\lambda\to\infty} f'(z,m,\lambda) &= 1 \ , \\ \lim_{\lambda\to\lambda_{\min}} f'(z,m,\lambda) &= -\infty \ , \end{aligned} \tag{31}$$

the saddle-function f must have an odd number of extrema. Our numerical analysis shows that it has exactly one minimum. When this minimum lies above zero, the saddle-point equations have no solutions. When the minimum lies below zero the saddle-point equation $f(z,m,\lambda) = 0$ has two solutions. Only the largest of these two solutions, however, is physical since at the smallest one $f'(z,m,\lambda) = 1/\rho < 0$. Then we have exactly one physical solution of the saddle-point equations.

In Fig.1 we plot the critical line in parameter space below which there exists one solution to the saddle-point equations for $D = 3$. We choose to plot this line as a function of the parameters m and $k_BT/r = 1/2z$, where T is the temperature.

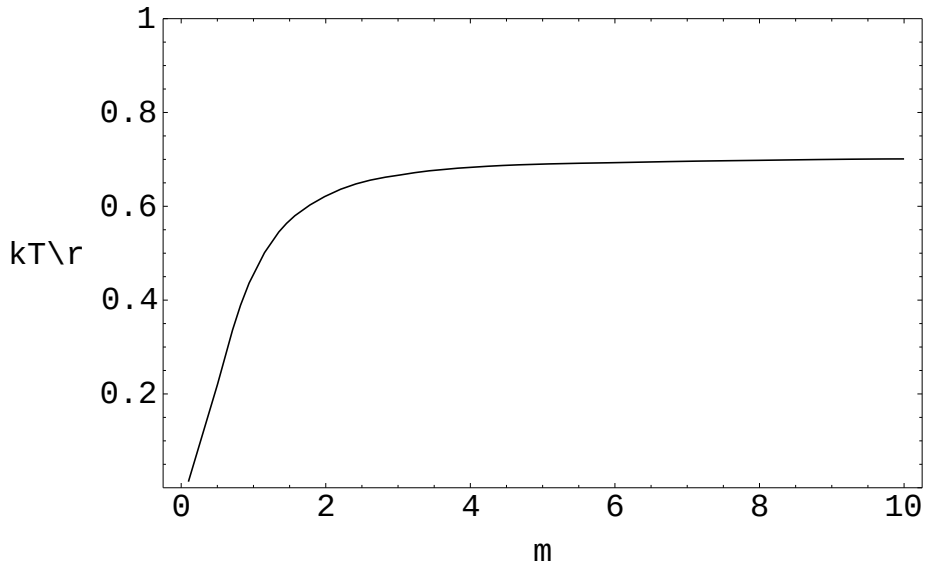

Fig. 1: Critical line for roughening transition at large-D.

When the critical line is approached from below, the minimum of the saddle-function $f(z,m,\lambda)$ goes to zero, so that $\lambda \to \lambda^*$ which is the solution of the equation $f'(z,m,\lambda^*) = 0$. Using (27), we conclude that

$$\lim_{(z,m)\to(z,m)_{\rm cr}} \rho \ = \infty \ . \tag{32}$$

Thus, when approaching to the critical line, the *intrinsic area* of the surface *diverges* whereas the renormalized tension remains finite. As far as the phase transition with respect to T or $1/r$ is concerned, this property is also found in the model with positive stiffness [8]. In the low-temperature phase of that model, however, there are short-range correlations between normals to the surface, indicating roughness [4,8]. As we show below, the situation is totally different in our model, as a consequence of the negative stiffness. Note that our model reproduces correctly the positive slope of the critical line in (T, n)-space, typical of ionic surfactants [10].

The geometric aspects of the surface are embodied in the correlation function

$$g_{ab}(\xi - \xi') = \langle \partial_a \phi^i(\xi)\ \partial_b \phi^i(\xi') \rangle\ , \tag{33}$$

for the normal components of the tangent vectors. This is immediately obtained from (21) as

$$g_{ab}(\xi - \xi') = \delta_{ab}\ \frac{1}{(2\pi)^2}\ \int d^2p\ \frac{1}{2\left[\lambda + W\left(z, m, p^2\right)\right]}\ \ \mathrm{e}^{i\sqrt{\rho}p(\xi - \xi')}\ , \tag{34}$$

where $W\left(z, m, p^2\right)$ is the Fourier transform of the interaction (15). This correlation function can be rewritten as

$$g_{ab}(\xi - \xi') = \delta_{ab}\ \frac{1}{(2\pi)^2}\ \int d^2p\ \left\{ \frac{1}{2\lambda} + \frac{z}{|\lambda|\left(\alpha - 2s\frac{|\lambda|}{z}p^2\right)} \right\}\ \ \mathrm{e}^{i\sqrt{\rho}p(\xi - \xi')}\ , \tag{35}$$

where we have used the fact that the solution for the saddle-point value of λ is negative. The dominant large-distance behaviour of this correlation function is given by:

$$g_{ab}(\xi - \xi') \simeq \delta_{ab}\ \frac{z^2}{8s\lambda^2}\ \sqrt{\frac{2}{\pi\sqrt{\frac{z\alpha\rho}{2s|\lambda|}}|\xi - \xi'|}}\ \sin\left(\sqrt{\frac{z\alpha\rho}{2s|\lambda|}}|\xi - \xi'| - \frac{\pi}{4}\right)\ . \tag{36}$$

It is easiest to check this result by computing backwards its Fourier transform. To this end it is useful to recognize that (36) represents the asymptotic behaviour of the von Neumann function $(z/8s\lambda^2)N_0\left(\sqrt{z\alpha\rho/2s|\lambda|}|\xi - \xi'|\right)$. Computing the two-dimensional Fourier transform of this function one finds [22] $z/|\lambda|(\alpha - 2s(|\lambda|/z)p^2)$ for all $0 < p < \sqrt{z\alpha/2s|\lambda|}$ i.e. one reproduces exactly the correlation (34) in momentum space.

The correlation function (36) is valid up to a large infrared cutoff R such that (1/R) provides a regulator for the pole at $p = \sqrt{z\alpha/2s|\lambda|}$) in (35). So, we have *long-range* order in the sense that there is no finite correlation length and $\int d^2\sigma\ g_{aa}(\xi)$ diverges like $\sqrt{R}$.

The oscillatory behaviour is due to the negative stiffness: it indicates that the surface fluctuates around the reference plane with an intrinsic wavelength

$$\ell \propto \sqrt{\frac{s|\lambda|}{z\alpha}} \ . \tag{37}$$

These long-range correlations are thus of a generalized antiferromagnetic type, with domains of orientational order of size ℓ. This is a consequence of the frustrated antiferromagnetic interaction with negative stiffness [18].

Contrary to models with a positive stiffness, our model predicts the existence of a smooth surface phase with long-range correlations for small T/r and large m. Notice that, due to (32) the correlations $\langle \partial_a \phi^i(\xi)\ \partial_b \phi^i(\xi') \rangle$ vanish on the critical line.te

We don't know the exact nature of the *roughening transition* occuring on the critical line depicted in fig. 1. The diverging intrinsic area and the vanishing correlators, however, indicate for certain that the rough surface is a fractal object. Note, however, that it is not of the type expected for a crumpled surface, where we should have a homogeneous solution to the saddle-point equations, while the correlation functions (34) would be short-range. It is not clear to us whether a generic solution of the saddle-point equations, not restricted by the ansatz (20), exists beyond the critical line. Such a solution would correspond to a fractal phase where the surface develops fingers and bumps. Also, it is not clear whether the large-D transition survives the corrections to D=3. If not, the surfaces would always be in the smooth, perturbative phase. In any case, we have shown how dipole dominance provides a viable mechanism for large, smooth surfactant interfaces. In addition, we have shown how a non-local interaction with negative stiffness leads generically to long-range, periodic correlation functions. Thus, our model might be of relevance for the recently observed [23] "egg carton" superstructure of membranes, although there are also other possible mechanisms to explain these [24].

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